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# Influence of sputter pressure on the texture and magnetic properties of barium ferrite thin films

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## Abstract

M-type barium ferrite thin films were deposited onto sapphire (001) substrates by radio frequency magnetron sputtering. Columnar grains were formed on the surface of the films under lower sputter pressure with bubble magnetic domains, which is originated from high magnetocrystalline anisotropy of the film. Magnetic hysteresis loops agree with the change of the morphology, where in-plane and out-of-plane loops are quite different of the samples prepared under high sputtering pressures, while they are almost identical under low pressures. The influence of the sputtering pressure was understood by that with the increase of the pressure, resputtering of the film is hindered. Nucleation with c-axis normal to the film plane was enhanced. Thus samples prepared under

low pressure have more acicular crystallites which have c-axis parallel to the film plane.

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Keywords: barium ferrite; thin film; sputtering pressure; morphology; XRD; VSM

## 1. INTRODUCTION

Barium hexaferrites (BaM,  $\text{BaFe}_{12}\text{O}_{19}$ ) with magnetoplumbite structures are the second generation of permanent magnetic materials which were developed into market in 1950s [1]. Bulk materials are widely used in traditional electro-acoustic devices, motors, and electricity generators, etc. Thin films can be potentially used in high density magnetic recording media [2,3] due to their excellent chemical stability, high mechanical durability, and large uniaxial anisotropy. They were proposed to be the key materials for thin film non-reciprocal microwave devices [4]. Very recently thin films of BaM were integrated with piezoelectric films, so that these microwave devices can be tuned electrically or magnetically [5]. In these applications morphology of the films, such as the shape, size and orientation of the grains, greatly influences performances of the devices. It was shown that the morphologies of the films are largely depending on the preparing methods and experimental conditions. Films sputtered by Chen et al. [6] consist of needle like grains. Thin films deposited by radio frequency diode sputtering show grains with irregular shape [7]. Using facing targets sputtering apparatus, films deposited on  $\text{SiO}_x/\text{Si}$  show needle and grounded grains

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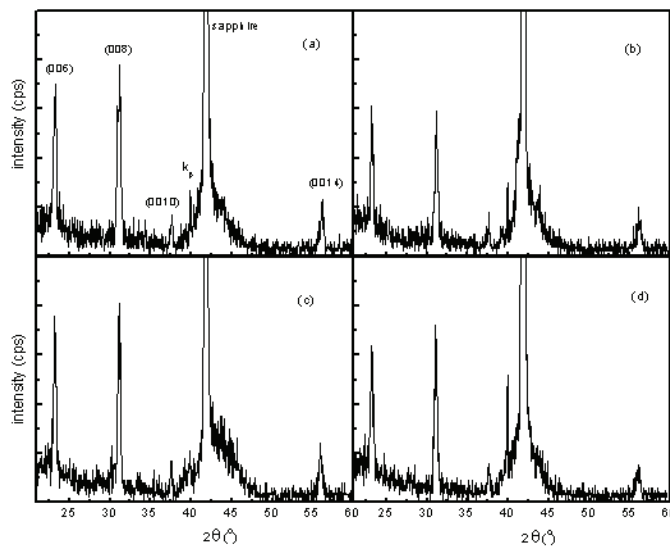
depending on the substrate temperature [8]. In this work BaM thin films were prepared by RF magnetron sputtering. The sputter pressure was used to modify the morphology of the film and its influence to the magnetic properties were discussed.

## 2. EXPERIMENTAL PROCEDURE

A buffer layer of BaM about 20 nm was sputtered at 2.0 Pa with base pressure of  $5.0 \times 10^{-4}$  Pa using Ar gases (purity of 4N) onto (001)-sapphire substrate. The samples were annealed at 1000 °C for 3 hours. Then the films were sputtered under different gas pressures: 0.2, 0.5, 0.8, and 1.0 Pa for 1 hour to reach thickness about 2  $\mu$ m. The films were annealed at 1000 °C for 3 hours in a tube furnace with two open terminals. A four-circle Bede D1 x-ray diffraction system with Cu-K $\alpha$  radiation (40 kV and 30 mA) was used to identify the phases and its texture. The morphology and magnetic structures were measured by scanning probe microscopy (SPA-300HV). Magnetic properties of the films were measured at room temperature using a vibrating sample magnetometer (VSM, Rikon Denshi) with a maximum of applied field of 2 T.

## 3. RESULTS AND DISCUSSIONS

The XRD diffraction pattern of the 2 $\theta$ -scan from the different samples were shown in Fig. 1. The (00 $l$ ) peaks are obvious in the figure which means that the films show  $c$ -axis normal to the film plane. However, they do not show noticeable differences among them when the sputtering pressure was varied. Phi-scan of the (114) peak clearly shows the six-fold symmetry of the crystallites as shown in Fig. 2 as indicated by the six peaks positioned every 60°. Dispersion of the  $c$ -axis was characterized by rocking curves of the (008) peak as shown in Fig. 3. The full width at half maximum (FWHM) are between 0.7°~0.9° indicating good  $c$ -axis texture of the films. Samples prepared at higher pressures show slightly narrower width than those at lower pressures. This was mainly due to the slower deposition rate at higher pressures.



**Fig. 1.** 2 $\theta$ -scans of the films. Films prepared under different pressures: (a) 0.2 Pa, (b) 0.5 Pa (c) 0.8 Pa and (d) 1.0 Pa.

Film surface morphology and magnetic structures are shown in Fig.4 as obtained by AFM and MFM. Bubble domains are visible which are because the magnetic moment in the bright (dark) regions is pointing up (down). The magnetic structures of the samples are quite similar as shown in Fig. 4(A), (B) and (C). However the morphology of the film shown in (a), (b) and (c) in the same figure is quite different. The samples prepared under lower pressure

have more needle-like crystallites. This crystallites with this shape were formed when the *c*-axis of the hexagonal platelet is not normal the substrate. This needle-like crystallites can be formed readily in all the *c*-axis in-plane textured specimens. For example, this kind of morphology transition was also observed in hexagonal AlN films [9]. However, these crystallites are invisible to XRD measurement because of their limited amount. At the same time, the *c*-axis points randomly in the plane, so coherent scattering of the X-ray is lacking. In this case, samples which have only (00*l*) reflection peaks have needle-like crystallites under microscopy. These diffraction peaks come from the massive crystallites with *c*-axis normal to the film plane. Reasons for the needle-like grains are still under debate. On sapphire substrate, fine grain sizes of the primary Ba-ferrite phase are obtained during crystallization, while on a SiO<sub>2</sub> substrate, amorphous Ba-ferrite film is naturally crystallized into the abnormal needle-like grains. It was suggested that the secondary  $\alpha$ -Fe<sub>2</sub>O<sub>3</sub> phase act as a useful inhibitor to abnormal grain growth in Ba-ferrite films by pinning the grain boundary movement in the later crystallization stage [10]. In sputtering, facing target sputtering produces films with *c*-axis normal to the film plane while normal sputtering produces films with *c*-axis parallel to the film plane with needle-like crystals. It may be because of re-sputtering of the film by the ions with high energy [11] in the normal sputtering configurations. Resputtering effect may be the reason for the larger amount of needle-like grains in samples prepared at lower pressure. In our experiments, the lower the sputtering pressure is, the more intensive the resputtering is.

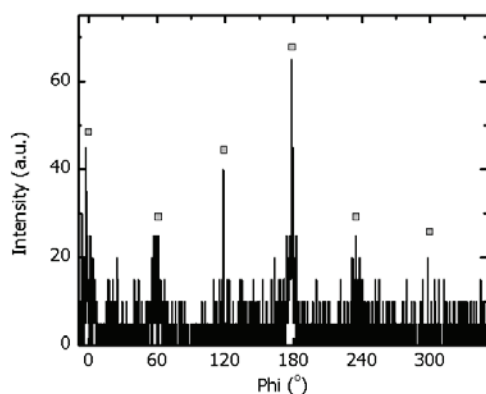


Fig. 2. Phi-scans of the film prepared at 0.5 Pa.

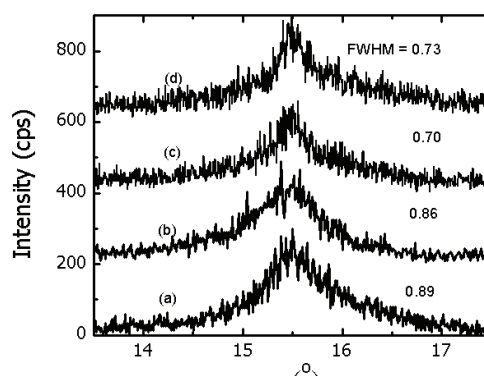


Fig. 3. Rocking curves obtained from the samples prepared under different pressures (a)0.2 Pa (b)0.5 Pa, (c)0.8 Pa and (d)1.0 Pa, respectively.

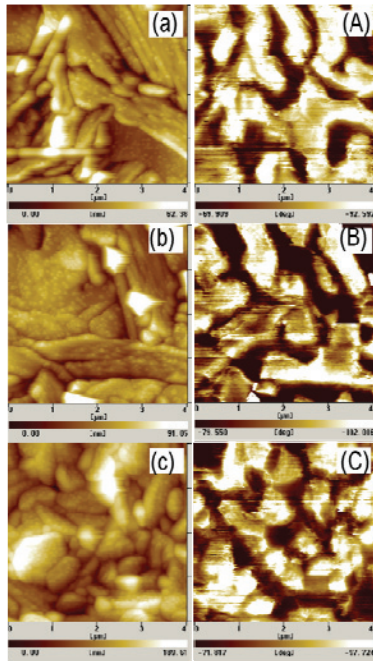
Magnetic hysteresis loops measured at room temperature show different shapes between in- and out-of-plane among the samples as shown in Fig. 5. The films prepared at 0.2 Pa show almost isotropic behaviors in the directions in- and out-of-plane as in Fig. 5(a). Films prepared under higher pressures show clear anisotropy (seeing Fig.5 (b)(c)). Fairly wide in-plane loops were observed in (a) indicating quite amount of in-plane *c*-axis orientation. In the low sputtering pressure region, the loops are almost isotropic. We also observed that in samples prepared at low pressure do not saturate in the normal direction at 2 T. This implies that a significant amount of crystallites have *c*-axis in the film plane, which are the needle-like grains. in both direction. Samples prepared at higher pressure show saturation in both directions above 1.2 T. The decrease of the remanent ratio as observed in the figures in the plane is consistent with the decrease of the needle-like crystallites with the increase of sputtering pressure.

#### 4. CONCLUSIONS

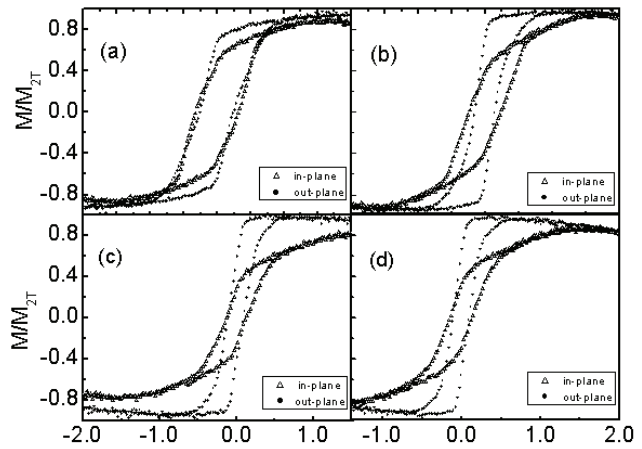
Barium hexa-ferrite thin films were fabricated by magnetron sputtering under different sputtering pressures. Surface morphology of the films changes from platelet-like to needle-like crystallites where the pressure was increased because of more bombardment of the films under lower pressures. The coercivity forces decreases monotonically with the sputtering pressure and the magnetic properties become more anisotropic, which is consistent with the decrease of the needle-like grains.

#### 5. ACKNOWLEDGEMENTS

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**Fig. 4.** AFM images (a,b, and c) of the samples and MFM images (A, B, and C) from the samples prepared under different pressures: A(a): 0.2 Pa, B(b): 0.5 Pa, and C(c): 1.0 Pa. The dark and bright regions in the MFM images correspond to the strength of the z-component of the magnetic moment.



**Fig. 5.** Magnetic hysteresis loops of the samples sputtered under different pressures: (a) 0.2 Pa, (b) 0.5 Pa, (c) 0.8 Pa and (d) 1.0 Pa.

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